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Performance of an integrated membrane pilot plant for wastewater reuse: case study of oil refinery plant in Indonesia

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ABSTRACT

This paper demonstrates the performance examination of an integrated membrane system (IMS) pilot plant having capacity of 250–300 L/h for oil refinery wastewater reuse of oil refinery plant in Indonesia. The IMS pilot plant consisted of a multimedia filter (MMF), an ultrafiltration (UF) unit, a reverse osmosis (RO) unit, and a membrane bioreactor (MBR) unit. The UF system was used as a pretreatment prior to RO, the RO unit was used to produce cooling water make-up and the MBR system was used for treating RO concentrate. The test results suggested that backwashing with a more frequent interval is needed to maintain stable flux of UF membrane. With the cleaning every 14 days, the resulting RO permeate was stable and able to fulfil the plant requirements of cooling water make-up. Treatment of RO concentrate using MBR unit resulted in permeate having relatively high chemical oxygen demand (COD). Further treatment by adsorption using granular activated carbon (GAC) proved that GAC was an effective method for reducing the COD level.

Keywords: Oil refinery wastewater; Cooling water; Integrated membrane system; Wastewater reuse

1. Introduction

Over the last two decades, membrane technologies have shown a significant growth in many industrial applications including water and wastewater treatments [1–6]. This is due to the more stringent regulations with respect to water quality and environment protection and the decreasing quality and quantity of available water resources. Among various wastewaters, oil refinery wastewater is one of the wastewater types, which is relatively difficult to be treated by conventional processes. In this case, membrane technologies have also been proposed as an alternative process for wastewater treating or recycling [3,4,7].

Typical sources of oil refinery wastewater are surface water runoff, cooling water, used process water, and sanitary wastewater [8]. The total amount of water used in petroleum refineries has been estimated to be 65–90 gallons of water per barrel of crude oil [9]. A significant amount is from the system for cooling purposes [10]. The environmental regulation forces the oil refinery plants to manage their wastewater. Two approaches can be performed to manage this wastewater namely minimization of wastewater via implementing clean production

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concept and treatment of wastewater for recycling or reuse purpose. Wastewater recycling or reuse reduces not only the amount of wastewater disposed to the environment but also the water consumption during production process.

For the purpose of wastewater reuse (e.g. for cooling water make-up), reverse osmosis (RO) has intensively been used in many industrial applications [7,11–13]. In principle, it can be said that RO is the powerful technique because the resulting permeate quality could be adjusted to meet the requirements needed. Nevertheless, fouling-deposition of (organic) material on RO membrane surface causing significant flux reduction—is the major limitation for these applications. Therefore, effective feed pretreatment should be conducted. So far, either activated carbon or low pressure membrane was widely used as a pretreatment of RO [14–17]. Ultrafiltration (UF) has shown as an effective method to remove macromolecule organic compounds such as humic acid and polysaccharides as well as microalgae [18–21]. The pretreatment method will depend strongly on the feed as well as the membrane characteristics. Another limitation of the RO is the resulting concentrate. This concentrate has very high concentration and in many cases it is not possible to dispose directly to the environment.

This paper presents the performance examination of an integrated membrane pilot plant, which generally consisted of an UF membrane, a RO unit, and a membrane bioreactor (MBR) system for treating oily wastewater of oil refinery plant in Indonesia (having capacity of 250–300 L/h). The multimedia filter (MMF) and UF units were used as pretreatments prior to RO and to produce cooling water make-up, whereas the MBR system was used for treating RO concentrate. The pilot experiment was performed to reduce the risk of process failure and to reduce cost, as it is less expensive than full-scale experiments. In addition, this pilot experiment was conducted to justify whether the process has potential to succeed on a full-scale basis.

2. Materials and methods

2.1. Membranes characteristics

The membranes characteristics used in this study are shown in Table 1. Hollow fiber membranes made from polysulfone were used for both the UF (pretreatment) and MBR system. Both membranes were manufactured by GDP Filter Company, Indonesia, in different module dimensions. While, a spiral wound thin film composite polyamide membrane manufactured by CSM Saehan Industries was used for the RO system.

2.2. Oil refinery wastewater characteristics as the feed

The wastewater used was from the outlet of Oil Impounding Basin (OIB) of oil refinery plant in Indonesia. The characteristics are presented in Table 2. Chemical oxygen demand (COD) in the effluent was within the range of 110–260 mg/L. However, it should be noted that COD in average was about 160 mg/L. It is important to mention that the wastewater contained high concentration of chlorides (177–509 mg/L) and total dissolved solids, TDS (605–1,053 mg/L).

2.3. Pilot plant system and experimental procedures

The schematic diagram of the integrated membrane system (IMS) pilot plant is depicted in Fig. 1. Three membrane processes, which are UF, RO, and MBR, were integrated into one sequence. The UF unit consisted of a UF membrane module, a feed pump, and a backwash pump (Lowara CEA series, CEA 70/3). The UF unit was also equipped with three pressure indicators to indicate pressure of feed inlet, concentrate outlet, and permeate outlet of the membrane module, three flow meters for monitoring flow rates of feed, permeate, and backwash. The RO unit consisted of a RO membrane module, a feed pump (CEA 70/3), and a high pressure pump (Lowara SV series, SV2 14). In order to monitor process conditions, the RO unit was also equipped with pressure indicators at inlet and outlet of the RO membrane module, and two flow meters for monitoring flow rate of permeate and concentrate. The MBR unit consisted of a bioreactor tank, two UF membrane module, a feed pump, and a backwash pump (CEA(M) 70/3). The MBR unit was also equipped with three pressure indicators (to indicate pressure of feed inlet, concentrate outlet, and backwash inlet) and two flow meters (to indicate flow rate of permeate line and backwash line). The granular activated carbon (GAC) unit was designed to achieve hydraulic loading rate of 1.5 m^3/m^2h and contact time of 12 min. A commercially available GAC from Calgon Corporation (FILTRASORB® 300) was used.

The presence of the UF unit as pre-treatment was expected to be able to produce the permeate stream as RO feed water having turbidity less than 0.5 NTU. Thereby, the possibility of (bio) fouling formation on RO membrane surface could be minimized. The RO membrane was used to treat further the UF permeate and was operated with the recovery target of 75%. The resulting RO concentrate was further processed using MBR system (in MBR system, UF membrane was used). The amount of the RO concentrate treated MBR was around 1,800 L/d. Finally, 50% of the MBR permeate was passed through a GAC filter.

Table 1
Characteristics of membrane modules

Parameter	Membranes		
	Ultrafiltration	Reverse osmosis	Membrane bioreactor
Model	S-640	RE4040-BE	S-240
Type	Hollow fiber	Spiral wound	Hollow fiber
Material	Polysulfone	Polyamide thin film composite	Polysulfone
Dimension	Dia. 6 in × L 40 in (0.1524 × 1.016 m)	Dia. 6 in × L 40 in (0.1524 × 1.016 m)	Dia. 2 in × L 40 in (0.0508 × 1.016 m)
Cut off/ rejection	MWCO 50 kDa	NaCl rejection 99,7%	MWCO 50 kDa
Membrane area	9.6 m^2	7.9 m^2	4.3 m^2

Table 2
Characteristics of oil refinery wastewater from OIB

Parameter	Range	Average	Standard deviation
pH	6.7–7.7	7.24	0.310
Sulfide, mg/L	0.04–0.08	0.05	0.009
COD, mg/L	110–260	163.48	37.216
Phenol, mg/L	0.5–0.7	0.58	0.077
Oil content, mg/L	4–6	5	1.000
Ammonia, mg/L	2.0–45.0	8.03	10.371
TDS, mg/L	605–1,053	877.24	211.460
Chloride, mg/L	177–509	341.67	97.669
Silica, mg/L	17.31–52.68	27.60	8.773
MLSS, mg/L	34–66	46.52	10.117

The experiment was conducted as the following procedures. Oil refinery wastewater from OIB was pumped into the strainer and MMF, respectively, to remove large (suspended) particles. The effluent of the MMF was collected in the UF feed tank and then was pumped into the UF membrane. UF permeate was collected in the RO feed tank as the feed of RO membrane. Before the RO feed water was pumped into the RO membrane module, sodium metabisulfite (SMBS, from PT Multi Kimia Raya, concentration of 97%) with dosing of 3 mg/L, antiscalant (PermaTreat® PC-510T from Nalco) with dosing of 3 mg/L, and H_2SO_4 (from PT Multi Kimia Raya, concentration of 98%) were added to remove chlorine, prevent scaling, and adjust the pH to 7.2, respectively. The RO unit was designed to operate under feed flow rate of 1.8 m^3/h and recovery of 75%. The RO concentrate (60 L/h) was treated in the MBR system, while the permeate of MBR system was further processed with GAC filters to improve the final effluent quality. In adition to treat further in MBR, the RO concentrate was also used as backwash liquid for the UF system with the backwash interval and duration were set automatically.

The MBR unit was designed for the aerobic system with the daily temperature of 28–37˚C, sludge retention time of 38 days, and hydraulic retention time of 24 h. The COD concentration in the feed was within the range 490–510 mg/L. The inoculum was obtained from the activated sludge, which was installed before the OIB unit. Therefore, an adaptation of the inoculum to the MBR was not necessary. However, the MBR evaluation was conducted after the process ran for one week. The MBR system was operated at the permeate flux of 15 ± 2 L/m^2 h. The performance of the MBR unit was examined in term of permeate quality. The GAC unit was integrated to the system (after MBR) to enhance the MBR permeate quality in case if necessary.

Membrane cleaning was investigated to obtain an effective cleaning procedure for the RO membrane. It was reported that ethylene diamine tetraacetic acid (EDTA) and strong acid show effective cleaning agents for restoring RO flux permeate [22–24]. However, in this case, citric acid was used instead of hydrochloric acid to avoid premature damage of the membrane [25]. The behavior of UF and RO permeates and their quality was investigated by measuring the flux and the concentration during 38 days. Citric acid and Na_4EDTA (both technical grade) were obtained from local Indonesia companies, i.e. PT Brataco and PT. Multi Kimia Raya, respectively. The RO membrane was firstly cleaned by using citric acid solution (1%) followed by EDTA solution (1%) and rinsing using water treated with RO. Membrane cleaning was performed by circulating cleaning solution with circulation rate was 130 L/min for each vessel for about 45 min.

In all experiments, the system was operated in continuous mode. The UF system was carried out in cross-flow filtration, where filtration and backwash

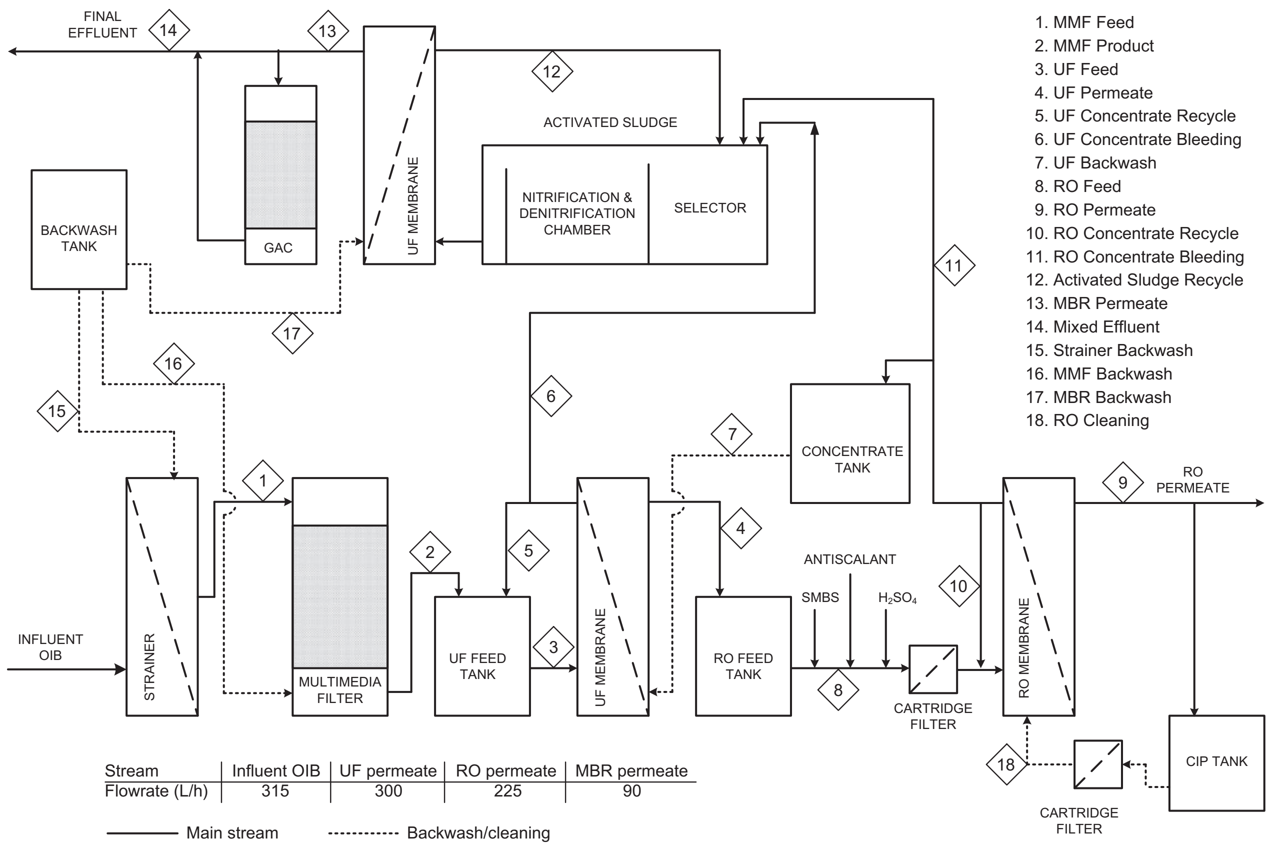

Fig. 1. Schematic diagram of the integrated membrane pilot plan for the treatment of oil refinery wastewater from OIB.

cycles ran alternately. Constant pressure mode with trans membrane pressure (TMP) of 0.7 bar was applied. The TMP was defined as follows:

$$\text{TMP} = \frac{P_{\text{in}} + P_{\text{out}}}{2} - P_{\text{perm}} \tag{1}$$

where P_{in} is the pressure enters into the membrane module through the feed side, P_{out} is the pressure out of the membrane module on the concentrate side, while P_{perm} is the pressure on the permeate side. The same pattern of operation was applied for the RO system, but at TMP of 12 bars.

3. Results and discussion

3.1. UF and RO membrane performance

3.1.1. UF flux behavior

In order to know the performance of the UF unit, the permeate flux behavior was firstly investigated during 38 days with backwash mode variation. The results are presented in Fig. 2. The results suggest that the backwash interval and duration had a significant effect on the permeate flux behavior (Fig. 2). Initially, backwash duration was determined based on our practical experiences (1 h for 3 min). In order to increase net product, we then tried prolonging the duration time to 24 h for 6 min and 6 h for 3 min (It should be noted that the shorter membrane cleaning interval the fewer net permeate product will be resulted). The permeate flux obtained with the backwash interval of 1 h and duration of 3 min was higher than the longer backwash interval (interval of 24 h operation and duration of 6 min as well as interval 6 h operation and duration of 3 min). This phenomenon indicates that the fouling tendency caused by physical interaction (foulant/particle deposition due to hydrodynamic pressure) was the dominant effect. With longer period of filtration, the adsorption should occur more significantly. It is important to note that membrane–solute interactions via adsorption should be difficult to be removed by backwashing.

3.1.2. RO flux behavior

Fig. 3 shows the RO permeate flow rate observed during 38 days. Systematic flux decline was observed

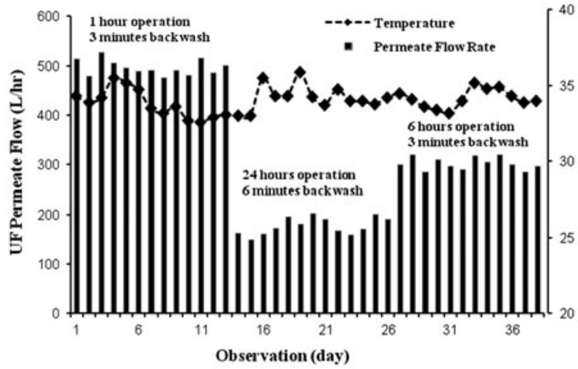

Fig. 2. Permeate flow rate of UF membrane.

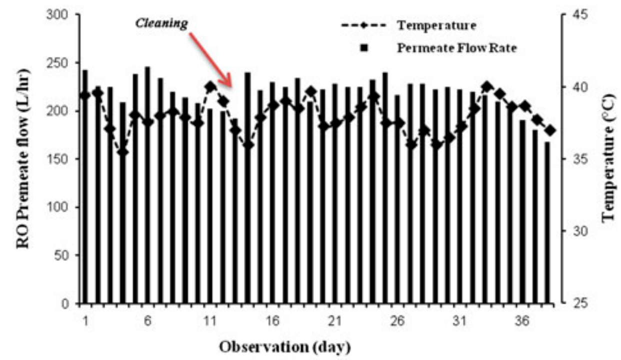

Fig. 3. Permeate flow rate of RO membrane.

after 6 days of operation. Our field experience suggested that we should clean the membrane when the resulting flux has already decreased by 20%. It was observed that after 13 days operation, the permeate flux was of 80% of the initial flux. Therefore, we cleaned the RO membrane. After membrane cleaning using EDTA and citric acid solution with duration of 40 min, the relative flux can be restored close to the initial condition. The flux decrease tendency in further operation was slightly slower and therefore performing the membrane cleaning every 14 days was still quite reasonable for maintaining the flux more than 80% of initial condition.

This cleaning duration is significantly faster than industrial application of RO membranes reported by Kucera [27]. Membranes with good pretreatment can expect to clean every about three months. However, different results were reported by Esfahani et al. [28]. Beside they found long duration in interval cleaning of RO, they also found that the RO membranes should be cleaned after 27 days of operation. To the best of our knowledge, cleaning duration is determined by the extent of fouling and scaling occured. Both processes are influenced by the membrane characteristic used, feed characteristic/pretreatment quality, and operation condition. Of course the cleaning duration in this experiment could be prolonged by installing a better pretreatment unit (e.g. lower pore size of UF membrane, disinfection, etc.). The better pretreatment used, the longer cleaning duration should be applied to RO membrane. In general, there will be a competition between investment and operational costs that should be considered.

3.1.3. RO permeate quality

Table 3 shows the characteristics of RO permeate. All parameters were lower than the requirements of the cooling water and therefore it was able to fulfil the plant requirements for cooling water make-up. Thus, this RO permeate can be directly used as an attempt to reduce water consumption in oil refinery plants as long as the feed quality can be maintained as in this experimental result.

3.2. MBR performance

The performance of MBR system (bioreactor and UF membrane) was observed from its permeate quality. The organic content expressed as COD in permeate was found within the range 110–150 mg/L, which is proportional to COD removal of 70–78%. Although these values fulfilled the Indonesian government regulation [27], disposing to the environment may have a risk due to interaction with other components and accumulation. To overcome this potential problem, the final COD was targeted to be less than half from the government standard (80 mg/L). Therefore, further treatment by adsorption using GAC was performed. The results showed that the activated carbon filters was found to be an effective method for reducing the COD content. This GAC could reduce the COD content from ~130 mg/L to ~65 mg/L.

Table 4 shows the product quality from the GAC unit. It is seen that the resulting product of GAC was compliant with the wastewater quality standards for refinery industry in accordance with the Regulation of Environment Ministry No. 19 Year 2010. Thus, the effluent from the GAC unit could be discharged directly.

3.3. Considerations of process implementation

In general, a selection of process for wastewater treatment is influenced by the feed wastewater quality and the targeted product/effluent. On the one hand, this advanced treatment using three different membranes needs high investment cost as well as

Table 3
RO permeate characteristics and cooling water make-up requirements

Parameter	Range	Plant cooling water make-up requirements [26]
pH	6.53–7.39	6.0–8.0
Hardness, mg/L	ND	Max. 70
Total alkalinity, meq/L	6.01–15.29	Max.50
Chloride, mg/L	2.9–20.0	Max. 50
Silica, mg/L	1.03–1.69	Max. 30
Ca hardness, mg/L	ND	Max.50
TDS, mg/L	18–73	Max. 200
Total Fe, mg/L	<0.02	Max. 0.3

ND: not detectable.

Table 4
MBR permeate characteristics enhanced with GAC system compared with the allowed discharge wastewater quality for oil refinery wastewater

Parameter	Range	Wastewater quality standards for the oil refinery industry [29]
pH	6.68–8.03	6–9
Ammonia, mg/L	<0.5	8
Phenol, mg/L	0.2–0.3	0.8
COD, mg/L	51–78	160
BOD, mg/L	4–8	80
Sulfide, mg/L	<0.01	0.5
Oil content, mg/L	1.0–3.2	20
Temperature, ℃	31–36	45

operation cost. On the other hand, by using this system an industry (here is oil refinery plant) can reuse their wastewater for both in process or other purposes. In addition, the quantity of the water source to be used and the wastewater to be disposed to the environment can be significantly reduced. Thus, the cost of production can be reduced significantly and the quality of wastewater can be increased. The application of this system in industry will be affected by environment legislation and the availability of water resource as well (price and quantity). In case that the conventional treatment can meet the environmental standard and the purpose of wastewater treatment is only to fulfil the effluent standards it may be not necessary to use this system. However, in case that the availability of the water can be a problem (cost, quantity, legislation) or the purpose of the treatment is to reuse the wastewater, this system can be an alternative technology. In this study, the availability of the water resources of oil refinery plant is very limited so that the company should use a treatment process to reuse their wastewater.

4. Conclusions

A pilot study of evaluation of an IMS for oil refinery wastewater reuse has been carried out. UF membrane permeate flux could be maintained by adjusting both the backwash interval and duration. RO membrane cleaning with EDTA and citric acid solution was able to restore the RO flux closed to the initial condition. The RO permeate quality fulfilled the requirements for cooling water make-up. The MBR permeate quality was increased by GAC filter, which was compliant with the wastewater quality standards for oil refinery industry in Indonesia. By using the results of this pilot experiment a process feasibility, process design, and process economic for full-scale can be determined.

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